SMC-TR-99-18

AEROSPACE REPORT NO.
TR-98(8555)-9

Quantum-Mechanical Interference Between Optical Transitions: The Effect of Laser Intensity Noise

15 May 1999

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19990603 163

Prepared for

SPACE AND MISSILE SYSTEMS CENTER
AIR FORCE MATERIEL COMMAND
2430 E. El Segundo Boulevard
Los Angeles Air Force Base, CA 90245

Engineering and Technology Group

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THE AEROSPACE CORPORATION
El Segundo, California

This report was submitted by The Aerospace Corporation, El Segundo, CA 90245-4691, under Contract No. F04701-93-C-0094 with the Space and Missile Systems Center, 2430 E. El Segundo Blvd., Suite 6037, Los Angeles AFB, CA 90245-4687. It was reviewed and approved for The Aerospace Corporation by R. P. Frueholz, Principal Director, Electronics Technology Center. Michael Zambrana was the project officer for the Mission-Oriented Investigation and Experimentation Program (MOIE) program.

This report has been reviewed by the Public Affairs Office (PAS) and is releasable to the National Technical Information Service (NTIS). At NTIS, it will be available to the general public, including foreign nationals.

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Michael S. Zambrana

Michael Zambrana
SMC/AXE

REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

1. AGENCY USE ONLY (*Leave blank*)

2. REPORT DATE
15 May 1999

3. REPORT TYPE AND DATES COVERED

4. TITLE AND SUBTITLE
Quantum-Mechanical Interference Between Optical Transitions:
The Effect of Laser Intensity Noise

5. FUNDING NUMBERS
F04701-93-C-0094

6. AUTHOR(S)
Camparo, J. C., The Aerospace Corporation; and Lambropoulos, P.,
Max-Planck Institute fur Quantenoptik

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
The Aerospace Corporation
Technology Operations
El Segundo, CA 90245-4691

8. PERFORMING ORGANIZATION REPORT NUMBER
TR-98(8555)-9

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
Space and Missile Systems Center
Air Force Materiel Command
2430 E. El Segundo Blvd.
Los Angeles Air Force Base, CA 90245

10. SPONSORING/MONITORING AGENCY REPORT NUMBER
SMC-TR-99-18

11. SUPPLEMENTARY NOTES

12a. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release; distribution unlimited

12b. DISTRIBUTION CODE

13. ABSTRACT (*Maximum 200 words*)

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14. SUBJECT TERMS
Atomic clocks Spectroscopy
Optical pumping

15. NUMBER OF PAGES
7

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
Unclassified	Unclassified	Unclassified	

NSN 7540-01-280-5500

Standard Form 298 (Rev. 2-89)
Prescribed by ANSI Std. Z39-18
298-102

Quantum-mechanical interference between optical transitions: The effect of laser intensity noise

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(Received 7 August 1998)

In a previous publication [Phys. Rev. A **55**, 552 (1997)] we considered the effect of laser phase fluctuations on three-photon–one-photon phase control of resonance-enhanced photoionization. Here we extend those studies by considering the role of laser intensity noise in addition to laser phase noise. While our results indicate that relative intensity fluctuations between the fundamental field and its third harmonic have a significant effect on control, the contrast between constructive and destructive interference is nonetheless two orders of magnitude, even under (reasonable) worst case situations. Consequently, neither laser intensity nor phase fluctuations appear to pose a serious impediment to the efficient phase control of atomic and molecular processes. [S1050-2947(99)00403-5]

PACS number(s): 32.80.Rm, 32.80.Qk

In $(3+1)$-photon phase control, an optical transition is excited via a three-photon and a one-photon pathway as illustrated in Fig. 1 for the case of Xe multiphoton ionization [1,2]. The field at the fundamental frequency ω_1 has a phase ϕ_1, while the third-harmonic field with frequency ω_3 has a phase ϕ_3. (Following common practice, the subscripts indicate the harmonic nature of the field and not the number of photons required for the transition.) These phases are controlled by passing the two fields through a dispersive medium of length L with indices of refraction n_1 and n_3 for the fundamental and third-harmonic fields, respectively. The rate of excitation is proportional to the square of the total transition amplitude and hence proportional to $\Omega_1^2[1+f^2+2f\cos(\theta)]$, where Ω_1 is the fundamental field's three-photon Rabi frequency, f is the ratio of the one-photon to three-photon Rabi frequency (i.e., $f\equiv\Omega_3/\Omega_1$), and the relative phase $\theta=\phi_3-3\phi_1$. (For the purposes of our work here, we have ignored any constant phase difference between the two quantum-mechanical paths since even in those cases where it might be nonzero it can be incorporated into the relative phase difference between the two fields.) As the relative phase difference between the two fields is varied, typically by changing the vapor pressure of the dispersive medium, the rate of excitation exhibits constructive and destructive interference. The maximum contrast between constructive and destructive interference occurs when f equals unity.

In a previous publication [3], we considered the influence of laser phase noise on this control process. Since the dispersive medium's propagation constant depends on frequency, the relative phase between the two fields is a stochastic quantity by virtue of the stochastic nature of the laser frequency: $\theta(t)=\theta_0+\delta\theta(t)$, where

$$\theta_0=\frac{3\bar{\omega}_1 L}{c}[n_3-n_1], \tag{1a}$$

$$\delta\theta(t)=\frac{3\,\delta\omega_1(t)L}{c}[n_3-n_1]. \tag{1b}$$

Here $\bar{\omega}_1$ and $\delta\omega_1(t)$ refer to the mean frequency and stochastic frequency fluctuation of the fundamental field, respectively. Somewhat surprisingly, our results indicated that orders of magnitude of contrast could be maintained even when employing phase diffusion fields (PDFs) with a linewidth of 3 cm^{-1}. Here we extend our previous investigation by considering fields with amplitude noise as well as phase noise.

One expects an amplitude noisy field to influence $(3+1)$-photon phase control since f (in addition to θ) may now be a stochastic quantity. Unfortunately, little detailed information is available regarding the relationship between a fundamental field's stochastic characteristics and those of a harmonic field generated within a nonlinear medium, specifically in the sense of coordinated experimental and theoretical studies mapping the stochastic characteristics of the fundamental field to the harmonic field. Of course, on general grounds it is known that a third-order process induced

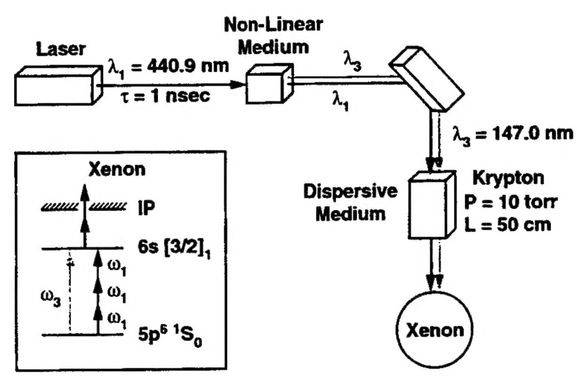

FIG. 1. Standard $(3+1)$-photon phase-control experiment. A fundamental field from a laser is tripled in a nonlinear medium and then both the fundamental and third harmonic pass through a dispersive medium. Since the refractive indices of the dispersive medium for the fundamental and third-harmonic fields are different, a relative phase difference between the two fields is created. Our numerical simulations consider $(3+1)$-photon phase control of xenon photoionization, where IP stands for ionization potential.

by a chaotic field should depend on a third-order intensity correlation function, which involves factors of 3!. However, the 3! term appearing in the third-harmonic field intensity should be similar to a 3! term appearing in the three-photon Rabi frequency, implying that f will nonetheless be constant, a conclusion borne out by results to be presented below.

To go beyond these general considerations, consider the nonlinear polarization equation, where the nonlinear medium's third-harmonic polarization P_3 generates the harmonic field E_3. Defining χ_3 as the nonlinear electric susceptibility, we have [4]

$$P_3(t)=\chi_3(\omega_1)\left(\frac{|E_1|}{2}\right)^3 \exp[-i3\omega_1 t]+\text{c.c.}, \tag{2a}$$

where [5]

$$\chi_3(\omega_1)\approx \sum_{n,m,q} \frac{\langle g|r|q\rangle\langle q|r|m\rangle\langle m|r|n\rangle\langle n|r|g\rangle}{(\omega_{qg}-3\omega_1)(\omega_{mg}-2\omega_1)(\omega_{ng}-\omega_1)}, \tag{2b}$$

and $\Omega_3\sim\chi_3(\omega_1)$. In Eq. (2b), $|g\rangle$ is the ground state and $\omega_{ij}=(E_i-E_j)/\hbar$. Given the stochastic nature of ω_1, each of the resonance factors in the denominator of Eq. (2b) will fluctuate and this will give rise to fluctuations in χ_3 and hence the one-photon Rabi frequency. Typically, however, the fluctuations of χ_3 are dominated by only one of the factors, specifically the two-photon resonance term. Further, since the fundamental field's frequency fluctuations will likely be small in comparison to the resonance detuning, Eq. (2b) can be rewritten as

$$\chi_3(\omega_1)\approx \sum_{n,m,q} \frac{\langle g|r|q\rangle\langle q|r|m\rangle\langle m|r|n\rangle\langle n|r|g\rangle}{(\omega_{qg}-3\bar{\omega}_1)(\omega_{mg}-2\bar{\omega}_1)(\omega_{ng}-\bar{\omega}_1)} \times\left[1+\frac{2\delta\omega_1}{(\omega_{mg}-2\bar{\omega}_1)}\right]. \tag{3}$$

(Notice that the correlation between χ_3 and the laser frequency fluctuations can be either positive or negative, depending on the detuning of the laser from the two-photon resonance.) Writing χ_3 in terms of its average value $\bar{\chi}_3$ and a mean-zero stochastic quantity $\delta\chi_3(t)$, Eqs. (2a) and (3) indicate that f is subject to additive noise and that under optimal conditions (i.e., $\langle f\rangle=1$)

$$f(t)=\left(1+\frac{\delta\chi_3(t)}{\bar{\chi}_3}\right)=\left(1+\frac{2\delta\omega_1(t)}{\Delta_2}\right), \tag{4}$$

where Δ_2 is the two-photon detuning in the nonlinear medium. Thus the stochastic variations of both θ and f in (3+1)-photon phase control derive from the fundamental field's frequency fluctuations.

The fundamental field is described in terms of our standard model for stochastic laser characteristics [6]: $E_1(t)=E_0(1+\epsilon)\cos[(\bar{\omega}_1+\delta\omega_1)t]$, where

$$\langle\epsilon(t)\epsilon(t+\tau)\rangle=\frac{\gamma}{\omega_f}\exp[-\omega_f|\tau|] \tag{5a}$$

and

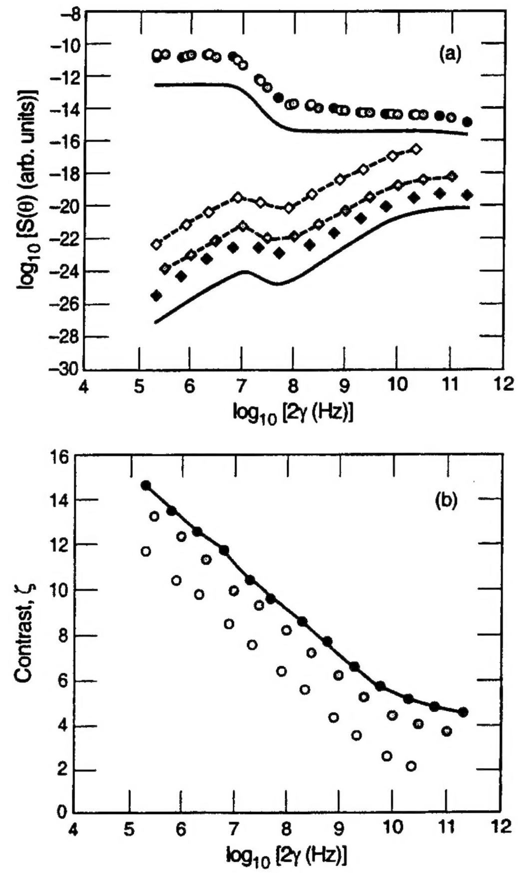

FIG. 2. (a) Signal amplitude versus laser linewidth. Circles correspond to constructive interference, while diamonds correspond to destructive interference: Filled black symbols correspond to $\Delta_2=\infty$, gray symbols correspond to $\Delta_2=100\,\text{cm}^{-1}$, and open symbols correspond to $\Delta_2=10\,\text{cm}^{-1}$. The solid lines correspond to previous results with a PDF, while the dashed lines are simply meant as an aid to guide the eye. (b) Contrast ζ versus laser linewidth. Symbol shading is as in (a) and again the solid line corresponds to previous PDF results.

$$\langle\delta\omega(t)\delta\omega(t+\tau)\rangle=\gamma\beta\exp[-\beta|\tau|]. \tag{5b}$$

Basically, γ defines the linewidth of the nearly Lorentzian line shape, β is a cutoff parameter for the line-shape wings, and ω_f is a bandwidth parameter associated with the fundamental field's amplitude fluctuations.

Our computational procedure is equivalent to that discussed previously [3]. We generate a realization of the fundamental field's frequency and amplitude fluctuations and then numerically integrate the relevant Xe density matrix equations for a 1-ns Gaussian pulse using a Runge-Kutta-Fehlberg technique [7], including now the third-harmonic field's additive noise. The signal that exhibits phase control is the total ionization $S(\theta_0)$ produced by the field during the pulse. Computing two ionization signals $S(0)$ and $S(\pi)$, we define the contrast of phase control as $\zeta=\log_{10}[S(0)/S(\pi)]$. For the results to be reported here, $\omega_f=3\gamma$ and $\beta=100\gamma$, indicating that the fundamental field was a near-chaotic field with an essentially Lorentzian line shape. The peak intensity of the fundamental field was $10^8\,\text{W/cm}^2$ (i.e., weak-field conditions) and the transition ac Stark shift was set to zero.

Figure 2(a) shows the logarithm of the signal amplitude as a function of the fundamental field's linewidth parameter (i.e., 2γ). Circles correspond to constructive interference signals, while diamonds correspond to destructive interference signals: For the black symbols $\Delta_2=\infty$ (i.e., no fluctuations in f), for the gray symbols $\Delta_2=100\,\mathrm{cm}^{-1}$, and for the open symbols $\Delta_2=10\,\mathrm{cm}^{-1}$. The solid lines correspond to our previous results with a PDF, while the dashed lines are simply meant as an aid to guide the eye. Considering for the moment the case of $\Delta_2=\infty$, laser intensity noise increases the signal amplitude dramatically for both constructive and destructive interference, as one might expect. Specifically, the signal enhancement factor is on the order of 50–70. (Note that for a nonresonant five-photon ionization process induced by a chaotic field, one would expect a 5! signal enhancement [8].) As additive noise for the harmonic field is increased, there is little change in the constructive interference signal amplitude, but a noticeable increase in the destructive interference signal. For example, in the case of $\Delta_2=10\,\mathrm{cm}^{-1}$ at $2\gamma=1$ MHz, the additive noise increases the destructive signal amplitude by more than three orders of magnitude even though $\delta f_{\mathrm{rms}}\sim 3\times10^{-5}$. This behavior is reflected in the contrast of phase control shown in Fig. 2(b), where the shading of the symbols is the same as in Fig. 2(a) and the solid line again corresponds to our previous results with a PDF.

Clearly, Fig. 2(b) demonstrates that a small amount of additive noise can have a significant influence on the contrast of phase control. However, even in the worst (reasonable) case of $\Delta_2=10\,\mathrm{cm}^{-1}$ and a field linewidth of approximately 1 cm^{-1}, the phase control contrast is two orders of magnitude. Thus, in an absolute sense phase control appears to be extremely tolerant of stochastic fields. The absolute degree of control predicted here, even in the worst case, is much better than what has yet been demonstrated experimentally, and from this we conclude that $(3+1)$-photon phase control is limited at present by processes other than those discussed here. For example, as noted by Chen and Elliot [9], overlap and focusing of the fundamental field and its first harmonic are extremely important for optimum phase control contrast. Given the present results, it is quite likely that these other experimental issues are the major processes limiting "orders of magnitude" $(3+1)$-photon phase control and not any inherent stochastic fluctuations of the laser field.

This work was supported under U.S. Air Force Contract No. FO4701-93-C-0094.

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